

AMENDMENTS TO SPECIFICATION

Page 1, prior to line 1, insert the section heading and text as follows:

CROSS-REFERENCE TO RELATED APPLICATIONS

This application is the national stage entry of International Application PCT/BE2004/000093 filed 25 June 2004, which claims priority from Belgian Application No. 2003/0411 filed 17 July 2003, the disclosures of both of which are hereby incorporated by reference.

BACKGROUND OF THE INVENTION

Page 2, lines 6-16:

FR-A-2784992 discloses an amylaceous adhesive composition for manufacturing corrugated paper board, which contains water, starch and an amount of two groups of active agents. The active agents serve the purpose of increasing the viscosity of the adhesive composition and of improving the wettability properties. The first active agent is a biodegradable soap, the second active agent is a sequestering agent for complexing alkaline earth metal ions such as calcium, magnesium, iron and others. Suitable sequestering agents include polycarboxylic acids and/or phosphonic acid and/or ~~polyacryl~~ polyacrylic acid, and or nitrilotriacetic acid or their alkaline salts and/or alkaline phosphates and mixtures thereof. The sequestering agent has the function of solubilising any fatty acids remaining in the soap.

Page 2, between lines 21 and 22 insert the section heading:

SUMMARY OF THE INVENTION

Page 2, line 29 – Page 3, line 2:

When analysing the problems occurring with the manufacturing of paper or card board, the inventor proposed to use an adhesive with a decreased penetration ability into the paper/card board, rather than an increased penetration ability as is taught by the prior art. In his opinion, decreasing adhesive penetration into the paper should allow saving on the amount of adhesive applied, limit the amount of water contacting the paper/card board and in that way limit the amount of water penetrating the paper/card board.

Page 3, lines 3 – 26:

The inventor has now found that a reduced card/paper penetration ability may be obtained with an adhesive which at the time of application has a lower viscosity than the known adhesive composition. The inventor thereby envisaged that with the reduced viscosity of the adhesive at the time of application, a smaller amount and a thinner layer of adhesive could be applied to the corrugations. With a thinner layer applied, the amount of water contacting and penetrating the paper/card board is reduced, thus reducing energy requirements for evaporating water that in the course of the lamination process had penetrated the corrugated paper board. The reduced water penetration also minimises the risk to deformation of the corrugated paper/card as well as of the flat liners. When further analysing the problems occurring with the manufacturing of paper or card board, the inventor concluded that the viscosity of the adhesive composition after application should be sufficiently high to ensure good adhesion.

To meet both requirements of a sufficiently low viscosity at the time of application of the adhesive and a sufficiently high viscosity at the time of effectuating the adhesion of the corrugated layer and the flat liner, the inventor proposed to add to the adhesive composition an additive as disclosed in the characterising portion of the first claim. The claimed additives show pseudo plastic properties and are capable of imparting this pseudo plastic behaviour to the adhesive composition containing them, even when present in a low concentration in the adhesive composition only. By incorporating the claimed additives, an adhesive composition can be obtained whose viscosity changes depending on the shear forces

that are applied to it. In particular, the adhesive composition containing the additive of the present invention has the effect that

Page 3, line 31 – Page 5, line 2:

- as soon as the adhesive composition has been applied to the corrugations, an almost instantaneous increase in the viscosity of the adhesive takes place, which prevents the adhesive from flowing away and ensures good adhesion. In this respect the inventor has found that initial viscosity of the additive is restored virtually immediately upon removal of the shear forces, as a consequence of which penetration of the adhesive into the card/paper board is minimised. The reduced adhesive penetration reduces water penetration into the paper/card board. This has the consequence that energy requirements for evaporating the penetrated water may be minimised, that a paper/card board with an improved strength may be obtained and that the tendency to lasting of the paper board may be reduced thus putting the paper board available for further processing after a shorter drying time. On the other hand, the viscosity is such that sufficient water penetration is ensured to minimise the risk to the formation of cracks in the paper/card board.

The inventor has further observed that with the claimed additives the viscosity of the adhesive composition may be reversibly increased and decreased. [[Thus,]] This had the advantage that any adhesive which has not be transferred from the applicator to the cardboard surface but remained on the applicator, may be recycled and re-used without this adversely affecting the adhesive strength. The inventor has further observed that the adhesive composition shows a better affinity for the card/paper board and provides an improved adhesion as well as improved printability of the surface as compared to prior art adhesive compositions. Prior art adhesive compositions used to control the viscosity of the adhesive composition by controlling the gelling temperature. However, gelling is an irreversible process[[. Once]] and once gelled, the adhesive composition cannot be re- used and loses its adhesive properties. With the present invention, the viscosity of the adhesive composition may be controlled independently

of the gelling process. The presence of the additive hardly affects the gelling properties of the composition.

The claimed additives have been found to show pseudo plastic behaviour and to impart this property to the adhesive composition containing them, even in case they are present in small amounts of for example 0.001- 5 wt. % or 0.001 - 1 wt. % only. As only a small amount is needed, the nature of the adhesive composition remains virtually unaffected, as well as its solids content and gelling properties, which is important when processing the composition.

The above described effects are particularly pronounced with an additive which is a homopolymer of acrylic acid, in particular a homopolymer of an $[[a]] \alpha$, $[[p]] \beta$ unsaturated monocarboxylic acrylic acid having 3-5 carbon atoms, which corresponds to formula $[[1]]$ I:

Page 5, line 10, please insert the following before the paragraph beginning "Homopolymers of acrylic acid and . . .":

The above described effects have also been observed with an additive which is a copolymer, in which the acrylic acid of formula I is co-polymerised with at least one alkyl acrylate of formula II:



In which R' is selected from the group of H, methyl, ethyl and R'' is a C10-C30 alkyl group, preferably a C10-C20 alkyl group, the copolymers being cross-linked with a cross-linking agent which is a polyfunctional vinylidene monomer containing at least two terminal methylene $\text{CH}_2=\text{C}$ groups as has been described above.

Representative acrylates of formula II include methylacrylate, ethylacrylate, propylacrylate, butylacrylate, methylmethacrylate, methylethacrylate, octylacrylate, laurylacrylate, stearylacrylate, behenyl acrylate, and the corresponding methacrylates. Suitable copolymers include copolymers of a mixtures of two or more of the afore

mentioned compounds with the acrylic acid of formula I. Other comonomers include acrylic nitriles, -olefinically unsaturated nitriles preferably those having 3-10 carbon atoms, for example acrylonitrile and methacrylonitrile; monoolefinically unsaturated acrylic amides, for example acrylamide and methacrylamide; N-alkylolamides of α,β -olefinically unsaturated carboxylic acids including those having 4-10 carbon atoms, preferably N-methylolmethacrylamide.

Page 5, lines 12 – 33:

An optimum viscosity ratio between a first situation in which shear forces are applied to the composition and a second situation in which shear forces are removed, is obtained with an additive in which the homopolymers or copolymers are cross-linked with a cross-linking agent which is a polyfunctional vinylidenealkylalkylene or a polyfunctional alkylalkylidene monomer containing at least two terminal methylene $\text{CH}_2=\text{C}$ groups, and having a molecular weight of between 0.05 - 100, preferably between 0.5 and 10, more preferably between 1 - 5 billion Dalton, the additive showing the property that upon application of shear forces, the viscosity decreases, the viscosity increasing instantly as the shear forces are removed.

Preferably use is made of a cross-linking agent which is a polyfunctional vinylidene monomer, more preferred a polyalkenyl polyether, in particular an allylpentaerythritol. Other suitable cross-linking agents include polyfunctional vinylidene monomers containing at least two terminal $\text{CH}_2=\text{C}$ groups, for example butadiene, isoprene, divinylbenzene, divinyl naphthalene, allyl acrylates, and the like. Preferred cross-linking agents are those containing an alkenyl group in which an olefinic double bond is attached to a terminal methylene group. Particularly preferred cross-linking agents include polyethers which contain on average two or more alkenyl ether groups per molecule. Other suitable cross-linking monomers include diallyl esters, dimethylallyl ethers, allyl or methallyl acrylates, acrylamides. Examples of those are allyl pentaerythritol, allyl sucrose, trimethylolpropane triacrylate, 1,6-hexanediol diacrylate, trimethylolpropane propane diallyl ether, pentaerythritol triacrylate and the like.

Page 6, please delete the paragraphs appearing on lines 1 – 22.

Page 6, line 23 – Page 7, line 19:

The adhesive composition of this invention presents the advantage of showing an increased cohesion, as a consequence of which the risk to cracking of the adhesive layer, once applied, may be reduced. The adhesive composition of this invention is particularly suitable for the manufacturing of the more recently developed corrugated paper/card boards, where there is an increasing tendency to reduce the width of the corrugations and increase the number of corrugations. In the recently developed corrugated boards, the corrugations may have a height of between 0.6 and 6.5 mm, the strength of the corrugations decreasing with increasing height. The adhesive composition of this invention is further particularly suitable for the manufacturing of the more luxurious, coated paper, which is usually coated with a coating agent that hampers evaporation of water. As with the present invention water penetration is reduced, the amount of water that needs to be evaporated is also reduced and the risk to formation of steam bubbles when evaporating the water is ~~minimised~~ minimized, even when used with coating agents showing a low water penetration.

Suitable commercially available products for use as the additive in the adhesive composition of this invention are available from Sigma Chemical Company, Rohm & Haas for example Acusol G[®], Noveon for example ~~Carbopol~~ Carbopol[®], from Wako Pure Chemical Industries of Japan, or from Allied Colloids of Great Britain for example Salcare Θ [®].

The additive which is incorporated into the adhesive composition of this invention will mostly be water soluble. It can be mixed as such with an adhesive composition in the solid state, or it may be added to an aqueous adhesive composition. To facilitate mixing, the additive is preferably diluted with a solid product before being added to the adhesive formulation. Suitable diluting agents are for example starch or inert inorganic oxides, for example siliciumdioxide, aluminium oxide, magnesium oxide, or mixed oxides of two or more of those. In that case the additive is mixed with the diluting agent before being added to the adhesive composition. The additive can be mixed in the form of a solid composition with a solid starch composition, or can be added to a liquid starch composition as a liquid composition. The

use of starch as a diluting agent for the additive assists in ~~minimising~~minimizing the risk to the formation of lumps upon mixing with the remainder of the adhesive composition.

Page 8, line 5 – Page 9, line 5:

The above described additive is suitable for use with a large variety of adhesive compositions, the nature of the adhesive composition not being critical to the invention. Suitable examples include adhesive compositions based on a polysaccharide adhesive, in particular a starch based adhesive although cellulose, or starch originating from peas, rice and potatoes are considered suitable as well. Thereby the starch may be a modified starch or the usual non-modified starch. The additive of the present invention is however also suitable for use with a polyvinylacetate based adhesive composition, although starch based adhesive compositions are preferred. Examples of suitable starch based adhesive compositions include so-called Stein-Hall adhesives, in which starch is present in both gelatinised and non-gelatinised form. A typical Stein-Hall adhesive for the manufacturing of corrugated board contains about 80 wt. % of water, about 15 wt. % of non-gelatinised starch, about 3 wt. % of gelatinised starch, about 0.55 wt. % of sodium hydroxide and about 0.4 wt. % of borax decahydrate although the weight ratios in which the different components are present, may vary. To this composition preferably about 0.1 wt. % of the additive is added.

Other suitable starch based adhesive compositions for use with the present invention contain 0.5-50 parts by weight of starch, between 0.01 – 2.5 parts by weight of alkali hydroxide, between 0.01-2 parts by weight of borax, 80-150 parts of water and 0.0008-10,25 parts of the additive. The gelatinised starch acts as the carrier and is an effective dispersing agent for the non-gelatinised starch. The non-gelatinised starch is caused to gelatinise on the warm paper surface, which involves a rapidly increasing viscosity. Similar examples of the above described adhesive formulation are the so-called no- carrier, Pristine and Minocar adhesives in which all the starch is in a swollen, but the non-gelatinised state. The starch may however also be at least partly pre- gelatinised or swollen. The inventors have now found that with the adhesive composition of this invention, the use of a carrier material can be dispensed with, which permits economizing on the starch. Thus, the amount of starch used in the adhesive

composition may be reduced with at least 2.5 wt. % with respect to the total weight of the composition, often even 4 or 5 wt. %. This embodiment presents the further advantage that a single phase adhesive composition is provided in stead of a two phase adhesive composition.

Furthermore, The starch used in the adhesive composition of this invention is selected from the generally commercially available starches, for example rice, wheat, corn, potato, tapioca or pea starch. The starch may be used in its natural form, or it may be physically, chemically or enzymatically modified. Physically modified starch is generally available in the form of roll-dried or extruded starch.

The adhesive composition may further contain an optical ~~clarification~~brightening agent, to allow controlling the quality of the adhesive ~~composition~~composition using UV irradiation.

Page 9, line 22 – Page 10, line 2:

In another embodiment, the polyacrylate homopolymer cross-linked with an allylpentaerythritol is first diluted with starch and an optical ~~clarification~~brightening agent in a weight ratio of 75-125, preferably 90-95 parts by weight of starch, 0,5-10, preferably 1-5 parts of optical ~~clarification~~brightening agent and 1-10, preferably 4-9 parts of polyacrylate. This mixture is then added to the above described adhesive composition in the above described ratio of between 0,001-5 wt % of additive with respect to the total weight of the composition.

The present invention also relates to a solid pre-mix which contains the adhesive composition of this invention as described above. The solid premix contains about 20-80, preferably 40-60 parts of gelatinised starch, about 5-50, preferably 10-30 parts of alkali, preferably caustic soda, about 200-750, preferably 350-550 parts of starch powder, about 1-25, preferably 5-15 parts of borax, and about 0.01-5, preferably 0.5 – 2.5 parts of the additive of the present invention described above. However, if so desired, the use of gelatinised starch may be dispensed with if so desired, as is disclosed above.

Page 10, lines 5 - 13:

The premix may be prepared by mixing the above described ingredients in the given amounts. The premix may for example be prepared by mixing 40-60 parts of starch with 400-600 parts of water, about 10- 20 parts of alkali, preferably caustic soda, about 350-550 parts of starch powder, about 5-15 parts of borax, heating the mixture to ~~gelatins~~gelatinse the starch. Thereafter, the water is evaporated to obtain a dry powder. To that powder about 0.5-2.5 parts of the additive of the present invention are added. The thus obtained premix needs only be mixed with a sufficient amount of water, for example about 500 - 700 parts of water to obtain the desired adhesive composition.

Page 11, line 4, insert the following paragraph before the paragraph beginning "Further preferred embodiments . . . ":

The present invention further relates to the use of the above described adhesive composition in the manufacturing of paper, according to which an amount of the adhesive composition is added to the paper pulp in the course of the paper production process.

BRIEF DESCRIPTION OF THE DRAWING

Page 11, between lines 6 and 7, insert the section heading:

DETAILED DESCRIPTION